The role of diagenisis in the hydrogeological stratification of carbonate aquifers: an example from the chalk at Fair Cross, Berkshire, UK

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The role of diagenesis in the hydrogeological stratification of carbonate aquifers: An example from the Chalk at Fair Cross, Berkshire, UK

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Abstract

Carbonate rocks form important aquifers in many parts of the world and in north-west Europe the Chalk is a primary source of potable water. When flushed with relatively fresh groundwaters, the Chalk may undergo significant diagenetic alteration at relatively shallow depths resulting in a physically and hydrogeochemically stratified aquifer. Diagenetic affects may have important implications for the effective exploitable thickness of the Chalk aquifer and for water quality. In order to assess the affects of diagenesis on the properties of carbonate aquifers, matrix porosity, permeability, pore water and rock chemistry profiles have been analysed for a 300 m deep borehole through the Chalk at the western end of the London Basin. An abrupt change in the matrix porosity profile at 155 mbgl indicates a change in dominant mode of historic diagenesis from mechanical compaction above 155 mbgl to predominantly pressure solution compaction below 155 mbgl. Pore water and rock chemistry profiles also change abruptly across this depth interval, suggesting that the present day hydrogeology is controlled by historic diagenetic trends. Below 155 mbgl, pore waters are relatively saline and there is no evidence for groundwater flow; above 155 mbgl pore waters are relatively fresh and geochemical evidence for incongruent carbonate dissolution indicates contemporary groundwater circulation. Possible physical and chemical evolution paths for the Chalk at Fair Cross are discussed. The results provide a hydrogeological context for other studies of the long-term response of carbonate aquifers to base-line changes in sea-level and pore water chemistry and also enable studies with relatively short time-frames or of localized phenomena to be placed in the broader context of the evolution of carbonate aquifers.

Introduction

The hydrogeological properties of the Chalk of the London Basin vary significantly with depth. Where it is unconfined, the upper 50 to 60 m of the saturated Chalk may effectively act as the aquifer (for example Water Resources Board 1972, Rushton & Chan 1976, Connorton & Reed 1978, Owen & Robinson 1978, Rushton *et al.* 1989). The high hydraulic conductivity of this zone, typically of the order of 10^{-3} ms^{-1} to 10^{-5} ms^{-1}, is principally due to the enlargement of the fracture network by dissolution processes (Price 1987). There is also limited evidence for the development of solution-enlarged fractures associated with a zone of enhanced hydraulic conductivity in the confined Chalk. Geophysical logs from the London Basin indicate that the majority of inflows in the confined Chalk are in the top 10 to 15 metres (Allen, Bloomfield & Robinson 1997). Below the zone of enhanced hydraulic conductivity, the hydrogeology of the Chalk is usually characterized by gradational changes in both physical and chemical properties. The average spacing of fractures is thought to increase with depth (Price *et al.* 1993), and it is assumed that fracture apertures will reduce in response to increased overburden (Brown & Scholz 1985). Consequently, as the fracture system becomes less hydraulically significant, it is envisaged that matrix porosity increasingly will influence solute movement. Onshore porosity profiles in the UK, particularly in the Upper Chalk in southern England where there is generally very limited lithological variation, are commonly characterized by a reduction in porosity with depth of burial (Edmunds *et al.* 1973, Barker 1994, Bloomfield *et al.* 1995). Changes in porosity with depth may contribute to gradual changes in water geochemistry away from the point of recharge (Edmunds *et al.* 1987, 1992) as both groundwater residence time and flow path length may influence water chemistry through the nature

and extent of incongruent reactions (Edmunds *et al.* 1987). Additionally, matrix porosity may influence the concentration profiles of relatively conservative major ion species, such as Cl^-, through diffusion gradients established between freshwater near the ground surface and water with marine affinities near the base of the Chalk (Bath & Edmunds 1981, Edmunds *et al.* 1987, 1992).

The purpose of this paper is to illustrate how carbonate burial diagenesis can influence both the physical and chemical evolution and the contemporary hydrogeology of carbonate aquifers. A specific example is used from the Chalk of the London Basin. Depth variations in hydrogeological characteristics are described using data from the Fair Cross borehole, Berkshire, a 248 m section through the confined Chalk at the western end of the London Basin. The physical and chemical data from Fair Cross are presented, following a brief discussion of the controls on carbonate diagenesis. The physical and hydrogeochemical stratification of the aquifer is described. Correlations are then established between the physical and chemical profiles and are interpreted and discussed in terms of the hydrogeological evolution of the aquifer.

Chalk diagenesis and hydrogeological stratification

There are two principal diagenetic processes that act to modify the pore structure of carbonate sediments such as the Chalk. These are mechanical compaction, the physical reorganisation of individual bioclastic fragments of the Chalk matrix into a progressively denser configuration, and pressure solution or solution transfer diagenesis, the reduction of matrix porosity through the dissolution and reprecipitation of minerals under conditions of non-hydrostatic stress (Neugebauer 1973, 1974, Scholle 1977, Scholle *et al.* 1983, Hancock 1993).

Mechanical compaction progresses with increased overburden and predominates during the early stages of burial diagenesis. During mechanical compaction, the sediment becomes denser through the deformation of coccoliths and their disaggregation into individual calcite laths, and through the reorientation and repacking of fabric components in the sediment. The degree of porosity reduction due to mechanical compaction is a function of local differential stresses, the strength, size, and shape of the matrix grains, and also the extent of prior cementation. Scholle *et al.* (1983) have suggested that, with a few hundred metres burial, mechanical compaction may result in the reduction of porosity from about 70% to less than 50% but, as porosity is reduced, an increasingly strong grain-supported fabric develops, mechanical grain re-orientation becomes difficult, and mechanical compaction becomes less efficient.

Pressure solution compaction becomes the primary mechanism for porosity reduction with a decrease in the effectiveness of mechanical compaction processes. Although limited pressure solution may occur during mechanical compaction, the onset of compaction predominantly by pressure solution occurs at matrix porosities in the range 45% to 35% (Scholle 1977, Scholle *et al.* 1983). Pressure solution is the process of dissolution of minerals under high non-hydrostatic stress and takes place at grain-grain contacts where pressure is greatest, because the solubility of calcite increases with increasing differential stress. Dissolved calcium carbonate migrates by grain-boundary diffusion, rather than by convective flow, down a chemical potential gradient to be precipitated at sites of relatively low stress (Bathurst 1971, Durney 1972). Pressure solution is controlled primarily by increasing overburden; however, the depth of onset of pressure solution and the rate of porosity reduction are also influenced by the chemistry of the interstitial water and the presence of clay minerals in the Chalk matrix.

Pressure solution in the Chalk is particularly sensitive to the chemistry of the formation water, and is enhanced if the original connate pore fluids have been flushed by relatively fresh, Mg-poor, waters (Neugebauer 1973, 1974, Scholle 1977), because magnesium can substitute for calcium in calcite, increasing its solubility. However, strong dependence of pressure solution on the concentration of Mg^{2+} in pore fluids has recently been questioned by Hancock (1993), who noted that Sr^{2+} can also substitute for calcium in low-magnesium calcite and may also modify the solubility of calcite. During freshwater flushing, changes in pore water chemistry are likely to be progressive (through slow flow and/or slow diffusive exchange). At any given time, the chemistry of the Chalk matrix may not be significantly out of equilibrium with increasingly fresh pore waters but the disequilibrium may be sufficient to enhance compaction by pressure solution. When dissolution and reprecipitation occur in the Chalk, the reaction is an incongruent reaction and pressure solution should lead to a purer calcium carbonate solid phase and to a more mineralized pore fluid. The initial clay content of the Chalk matrix is another important factor in controlling both the depth of onset of pressure solution and the rate of matrix porosity reduction by pressure solution. Small initial bed-to-bed variations in sediment composition can lead to major diagenetic variations during pressure solution of carbonates (Scholle 1977, Scholle *et al.* 1983), and clay-rich, carbonate-poor beds are particularly susceptible to pressure solution. Weyl (1959) has shown that the presence of clay minerals aids diffusional transport and this may explain in part why pressure solution is enhanced in Chalks with relatively high clay contents. Fine clay coatings on calcite laths may also inhibit reprecipitation of calcite in clay-rich beds.

The overall effect of interstitial water chemistry on porosity-depth trends and the transition from mechanical to pressure solution compaction as a function of porosity

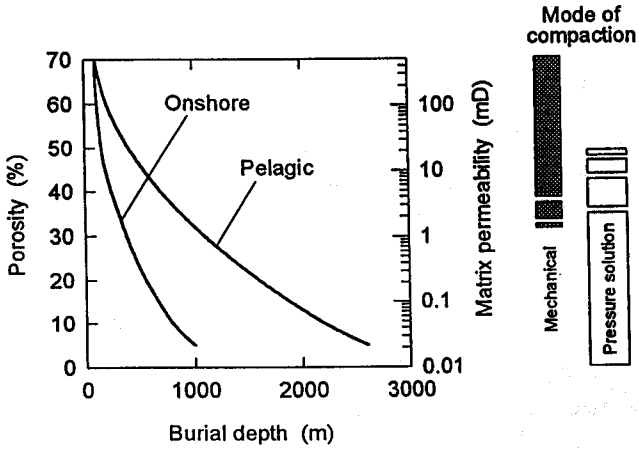

Fig. 1 *Schematic illustration of the variation in matrix porosity with burial depth for chalk containing saline pore waters, the 'pelagic' curve, and for chalks that have been flushed by fresh groundwater prior to burial, the 'onshore' curve. Approximate equivalent matrix permeabilities are also indicated. To the right of the graph the bar chart shows the dominant mode of chalk compaction as a function of matrix porosity. Note that there is a transition from dominantly mechanical compaction to dominantly pressure solution compaction at porosities in the range 45 to 35%.*

is illustrated schematically in Fig. 1. Scholle *et al.* (1983) described two end member trends in porosity reduction, denoted by the 'pelagic' and 'onshore' curves in Fig. 1. 'Pelagic' chalks with predominantly marine pore waters exhibit a smaller rate of porosity reduction with respect to depth of burial than 'onshore' chalks which have undergone flushing with freshwater. The figure shows that the transition between mechanical and pressure solution compaction may be expected at burial depths of 1000 m or more in 'pelagic' chalks, but may be as shallow as approximately 300 to 400 m in 'onshore' chalks.

Scholle *et al.* (1983) have shown that the porosity-depth profile which develops during pressure solution compaction may be expected to become increasingly heterogeneous as diagenesis proceeds. During pressure solution, carbonate material passes into solution in units containing relatively Mg-rich carbonates, clay-rich beds or finer-grained chalks; it is precipitated in adjacent beds as overgrowth cements, resulting in amplification of primary compositional variations in the sediment. Scholle (1977) refers to the process of preferential dissolution of selective beds, or 'donor units', and the preferential cementation of other beds, or 'recipient units', as 'diagenetic unmixing'. Garrison & Kennedy (1977) have described a range of pressure solution fabrics, including solution seams and flaser structures, or marl plexuses, small ellipsoidal bodies or lenses of relatively pure chalk surrounded by clay-rich solution seams; they have shown that these structures developed on the scale of centimetres to tens of centimetres during diagenetic unmixing by pressure solution. Under more extreme conditions stylolites may develop. These are the insoluble residue from a zone of highly localized pressure solution (Bathurst 1995).

Fair Cross study area

The Fair Cross Borehole was drilled in 1973 as part of a multi-disciplinary research programme into the hydrogeology of the Chalk of southern England. Water and rock chemistry data from the Fair Cross borehole have been published as part of a study of geochemical baseline conditions in the Chalk aquifer (Edmunds *et al.* 1987, 1992), the porosity-depth profile has been presented as part of a review of porosity trends in the Chalk of England (Bloomfield *et al.* 1995), and Morgan-Jones (1977) included data from Fair Cross in a description of the mineralogy of non-carbonate material from the Chalk of Berkshire and Oxfordshire. A detailed lithostratigraphic log and a range of geophysical logs for Fair Cross can also be found in Jones *et al.* (1997).

GEOLOGY

The Fair Cross Borehole (NGR SU 6974 6326) is situated approximately 10 km south-west of Reading, Berkshire, Fig. 2. The borehole penetrates 74 m of Palaeogene deposits (51 m of London Clay and 23 m of Reading Beds), 248 m of Chalk, and was terminated in the Upper Greensand at 329m total depth (Jones *et al.* 1997). The Chalk sequence at Fair Cross starts at 74 m below ground level (mbgl), which is 54.75 metres above OD, and below a Palaeogene erosion surface. The borehole is lined down to 95 mbgl, because the upper section of the Chalk is highly fractured and poorly consolidated. A simplified lithostratigraphic log, and calliper, gamma, micro-resistivity, and neutron porosity logs for Fair Cross are shown in Fig. 3. Fractures are present down to a depth of about 280 mbgl. These are present as infrequent zones of relatively intense fracturing at 146 mbgl and 238 mbgl, as joints, and as partings along marls. Two faults of unknown orientation and throw have been inferred from the core, one at 200 mbgl, at the top of the Chalk Rock, and a second at 215 mbgl within the New Pit Chalk. The joints are largely restricted to the 95 to 150 mbgl depth interval. Below approximately 150 mbgl, the most common fractures are those associated with marl partings. The marl partings commonly show well developed slickensides, and it is inferred that small lateral movements have occurred on many of these horizons.

HYDROGEOLOGY

The hydrogeology of the London Basin has been investigated extensively, and reviews can be found in Water Resources Board (1972) and in Allen, Bloomfield &

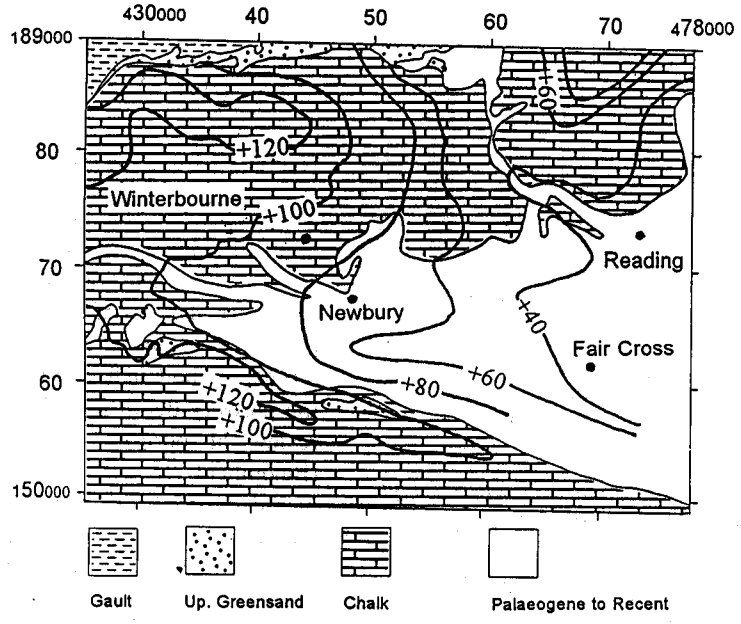

Fig. 2 *Sketch map showing the location of the study area at the western end of the London Basin and the positions of the Fair Cross and Winterbourne boreholes. The Chalk potentiometric surface is shown in metres relative to OD.*

Robinson (1997). The Chalk is the principal aquifer in the London Basin. It is overlain by the Reading Formation in the west of the Basin and underlain by the Upper Greensand and the Gault clay. The Reading Formation, fluviatile gravels and sands and alluvial clays, is generally in hydraulic continuity with the Chalk (Lloyd 1993) and provides leakage into the Chalk. Below the Chalk, the Upper Greensand acts as a minor aquifer and, below this, the Gault clay forms the basal aquiclude for the water bearing formations. At the western end of the Basin, recharge occurs along the Chilterns to the north, and through the exposed Chalk of the Berkshire and Marlborough Downs to the west and southwest.

Fig. 2 shows contours in metres on the potentiometric surface of the Chalk, and the present rest water level in the borehole is at approximately 30 maOD. There are no pumping test data for the Fair Cross borehole. However, Owen and Robinson (1978) reported transmissivity and storativity (T and S) in the ranges of 50 to 2500 m^2d^{-1} and 0.001 to 0.02, respectively, for the unconfined Chalk in the vicinity of the Lambourn and Pang rivers to the north and north-west of Fair Cross. In the confined Chalk at the western end of the London Basin, they reported T and S values in the ranges 270 to 450 m^2d^{-1} and 0.0002 to 0.00085 respectively.

Physical and chemical profiles

The porosity and gas permeability measurements presented and discussed in the following sections were obtained using 2.5 cm diameter plugs cut from the whole core. Porosity was determined using the liquid resaturation technique (Bloomfield *et al.* 1995), and gas permeability was measured using a steady state method at ambient conditions with nitrogen as the permeant (API 1960). Details of the analytical methods used in the determination of the major and minor elements can be found in Edmunds *et al.* (1987), and details of Mg^{2+} and Sr^{2+} whole rock determinations are given in Bath & Edmunds (1981).

POROSITY AND MATRIX PERMEABILITY

Fig. 4a illustrates the distribution of intergranular porosity with depth at Fair Cross and Fig. 4c is the corresponding normal probability plot. Fig. 4b shows the variation in matrix gas permeability with depth and Fig. 4d is a normal probability plot of the permeability data. Porosity ranges from 5.6% to 46.2% and there are two statistically discrete sub-populations, each approximating to a normal distribution. Samples with porosities >42% have a mean porosity of 43.6%, and an associated standard deviation of 1.0% (a variation of a similar order to the measurement error). Samples with porosities <42% have a mean porosity of 30.4%, with a standard deviation of 7.2%. Fig. 4a shows these two sub-populations correlate with two depth intervals. All samples below 155 mbgl have porosities <42%, and the majority of samples above 155 mbgl have porosities >42%. Fig. 4a shows that the transition between these two porosity regimes is abrupt and is of a similar order to the typical sample spacing (*ie.* less than 1.5 m). Fig. 5a and 5b present semivariograms for porosity data in the depth intervals 95 to 155 mbgl and 155 to

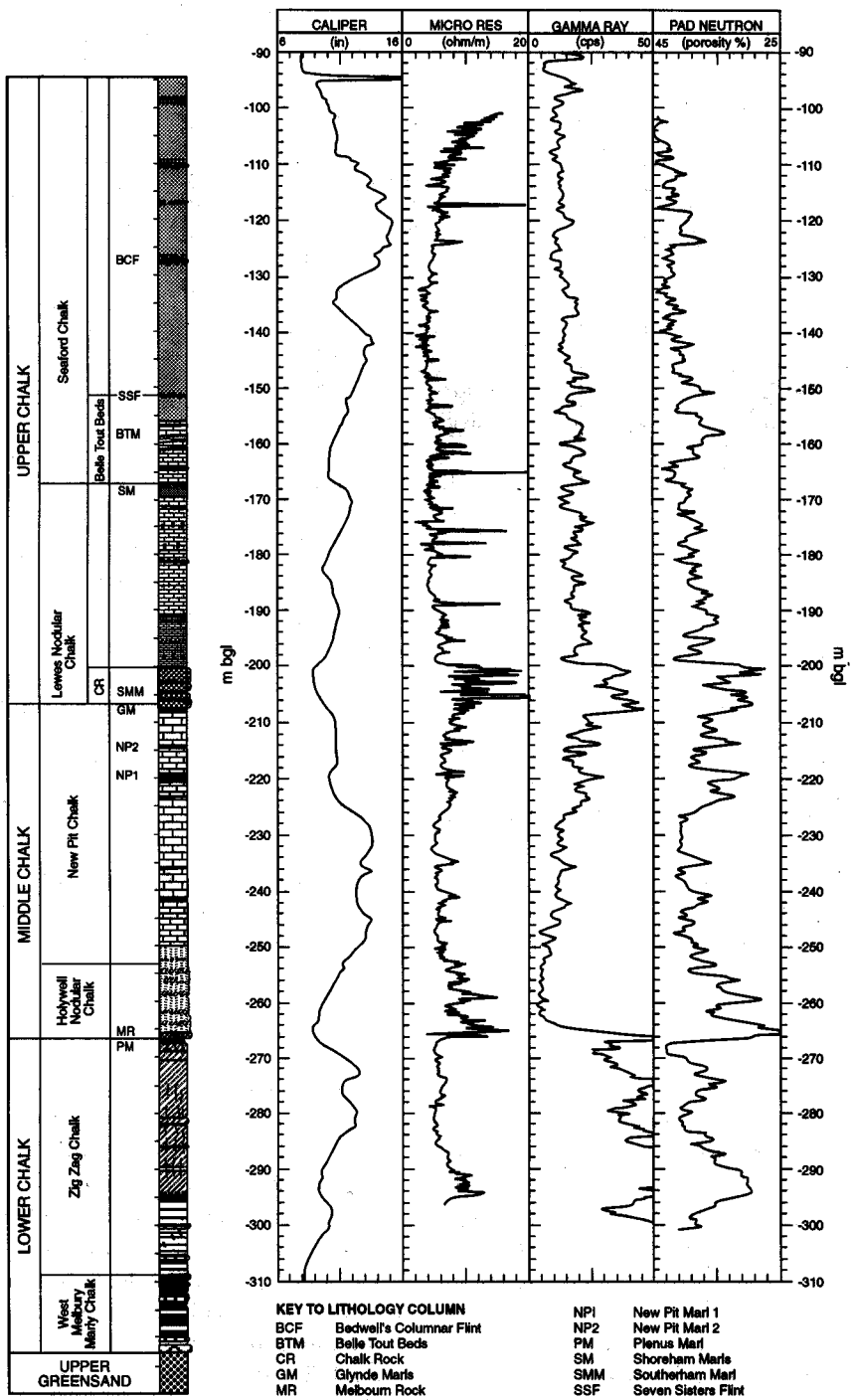

Fig. 3 *A simplified lithostratigraphic log, and calliper, gamma, micro-resistivity, and neutron porosity logs for the Chalk at Fair Cross. Depths are in metres relative to ground level.*

310 mbgl respectively. They show that, above 155 mbgl, there is no apparent correlation between sample porosities over correlation lengths greater than the average sampling interval but, below 155 mbgl, there is an apparent correlation between sample porosities up to a correlation length of approximately 24 m.

Matrix gas permeabilities range from 0.6 mD to 59 mD, equivalent to hydraulic conductivities in the range 10^{-5} to 10^{-2} ms^{-1}. Fig. 4b illustrates that there is a general reduction in matrix gas permeability with depth from about 10 mD at 95 mbgl to 0.05 mD at 320 mbgl. There are three approximately log-normal

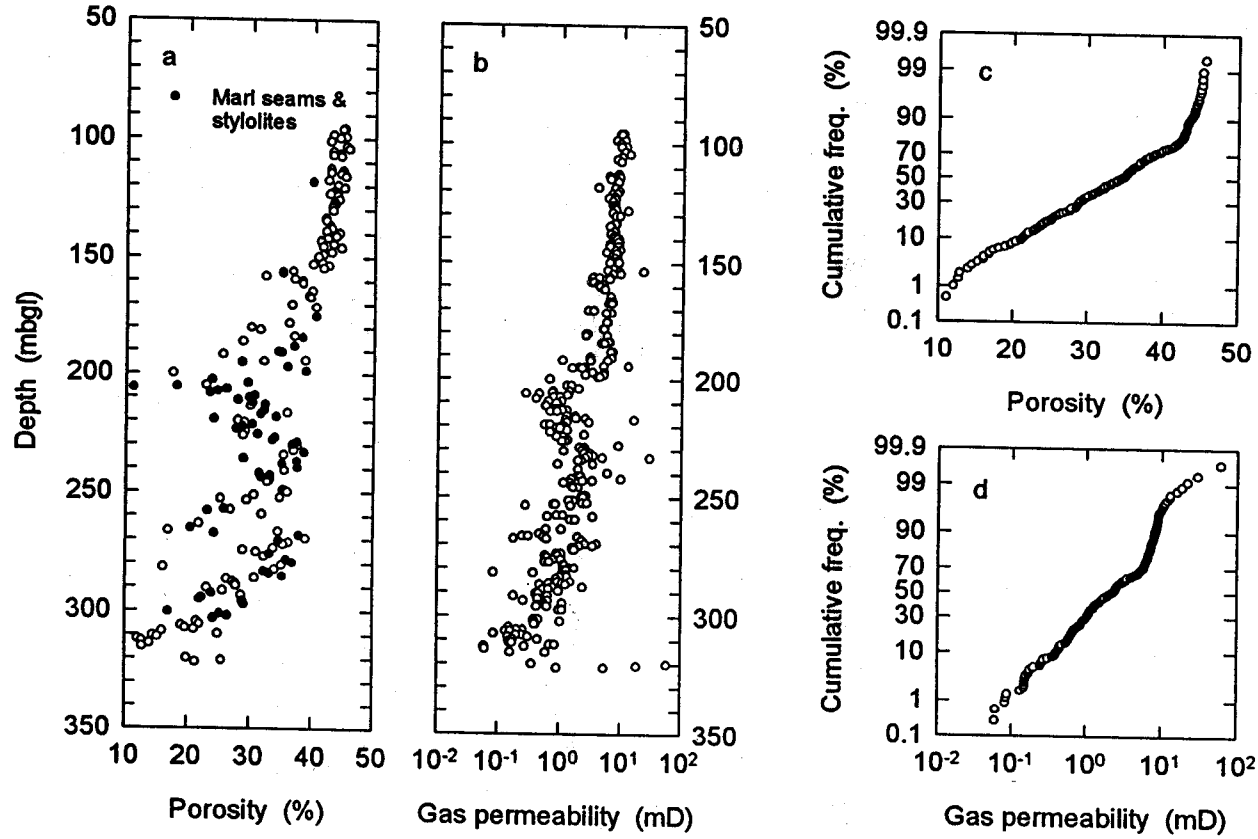

Fig. 4 *Illustration of (a.) matrix porosity, and (b.) matrix gas permeability depth trends at Fair Cross. Depths are given as metres below ground level. (c.) is a normal probability plot of the porosity data, and (d.) is a log-normal probability plot of the gas permeability data. In Figure 4a samples that contain marl seams and stylolites are denoted by closed circles.*

sub-populations within the gas permeability data (Fig. 4d). The limited number of samples which exhibit gas permeabilities >10 mD are associated with core plugs which contain continuous fractures; it is inferred that these values are not representative of matrix permeabilities (the fractures were either generated due to sample preparation procedures, or are partings along marl seams in horizontal samples). The second, statistically discrete, group of samples has matrix gas permeabilities in the range 6 mD to 10 mD with a geometric mean of 7.5 mD, and the third group of samples has permeabilities <6 mD with a geometric mean of 1.2 mD. These last two sub-populations can also be correlated with two discrete depth intervals analogous to the depth intervals defined by the porosity data.

INTERSTITIAL WATER AND WHOLE ROCK CHEMISTRY

Variations in interstitial water chemistry and whole rock chemistry with depth at Fair Cross are illustrated in Figs. 6 and 7. Figs. 6a and 6b show changes in the concentration of Cl^- and Na^+ in interstitial waters with depth and Fig. 6c is a cumulative frequency plot of the Na^+ data. Figs. 7a to 7d show the trends in Mg^{2+} and Sr^{2+} whole rock and interstitial water chemistry.

The pore water profiles in Figs. 6 and 7 show that there is an overall increase in salinity with depth. The Cl^- and Na^+ profiles have been described previously as exhibiting smooth increases with depth, with a stepped increase across the Chalk Rock at 200 mbgl (Edmunds *et al.* 1987, 1992). However, careful examination of Fig. 6 shows that there is a change in the slope of the Cl^- concentration profile at about 155 mbgl, and the Na^+ profile shows a change in slope at about 130 to 140 mbgl and at about 260 mbgl. Above 155 mbgl, the slope of the Cl^- concentration profile is -9.6 $mgl^{-1}m^{-1}$ but, below 155 mbgl, the slope approximately halves to -18.8 $mgl^{-1}m^{-1}$. The slope of the Na^+ concentration profile also approximately halves from -6.2 $mgl^{-1}m^{-1}$ between 95 and 140 mbgl, to -13.0 $mgl^{-1}m^{-1}$ between 140 and 260 mbgl. Below 260 mbgl, Na^+ concentrations are constant at approximately 1800 mgl^{-1}. These trends can be seen more clearly if the figures are viewed obliquely in the

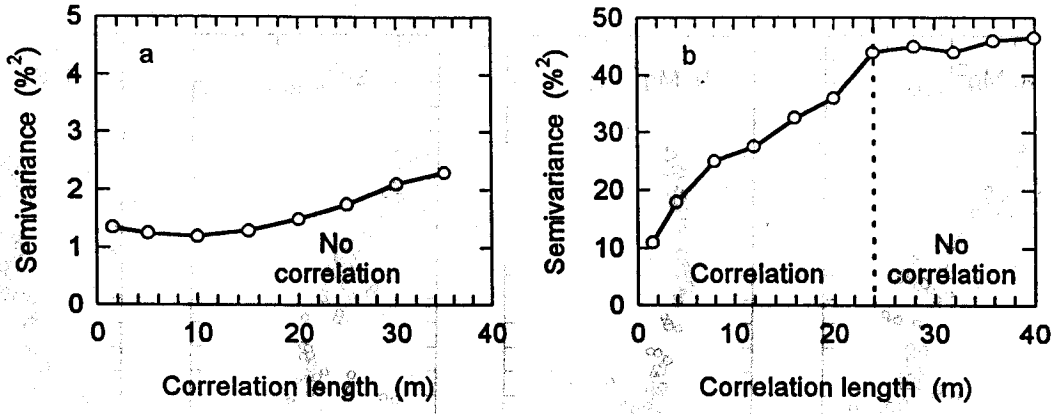

Fig. 5 *Semivariograms constructed for (a.) porosity data in the depth interval 95 to 155 mbgl, and (b.) porosity data in the depth interval 155 to 310 mbgl at Fair Cross. Figure 5 shows that there is essentially no spacial correlation between porosity measurements in the Chalk down to 155 mbgl, but above 155 mbgl there is an apparent correlation in sample porosities up to a length of approximately 24 metres.*

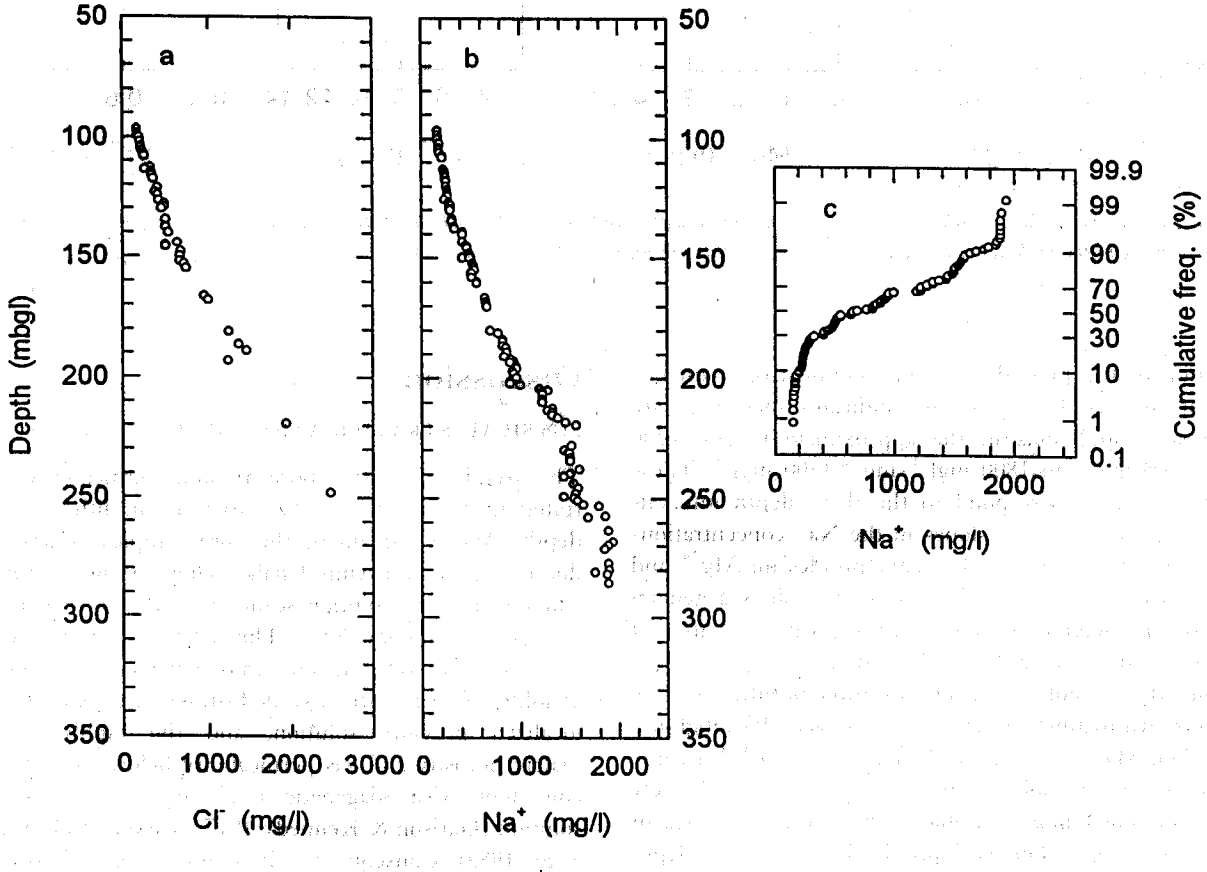

Fig. 6 *Illustration of the variation in (a.) Cl^- and (b.) Na^+ concentrations with depth at Fair Cross, where depths are in metres below ground level. (c.) is a normal probability plot of the Na^+ concentration data.*

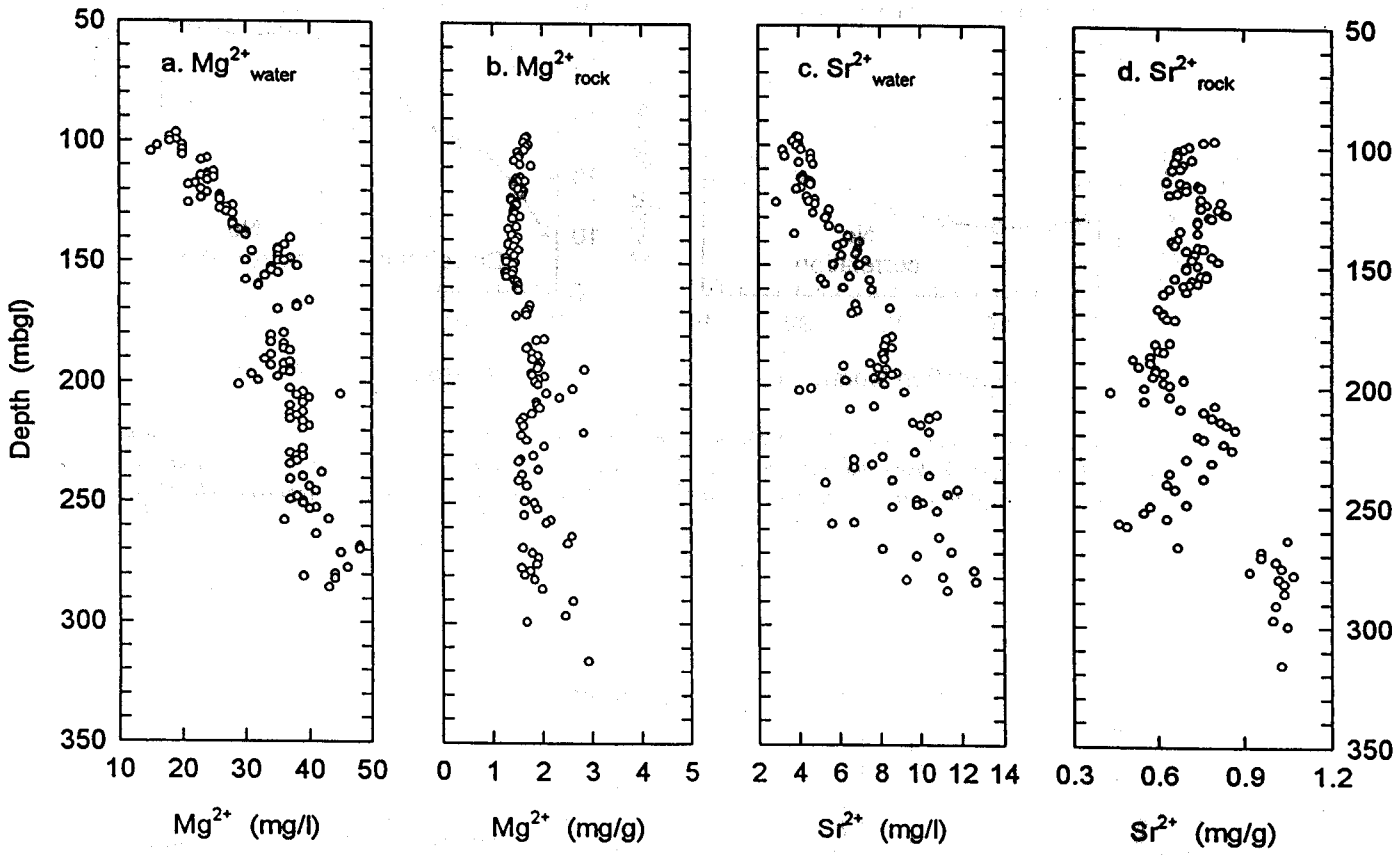

Fig. 7 *Illustration of the variation in (a.) magnesium pore water and (b.) whole rock chemistry, and (c.) strontium pore water and (d.) whole rock chemistry with depth at Fair Cross, where depths are in metres below ground level.*

direction of the plotted data. The cumulative frequency plot of the Na^+ data, Fig. 6c, indicates that there are three sub-populations in the approximate ranges <200 mgl^{-1}, 200 mgl^{-1} to 1800 mgl^{-1} and >1800 mgl^{-1}. These sub-populations correspond to the three depth intervals defined by the different slopes in the Na^+ concentration-depth profile. The water chemistry profiles for Mg^{2+} and Sr^{2+} concentrations, Fig. 7a and 7c, also show a general increase in mineralisation with depth, with the greatest increase in the interval 95 to 155 mbgl.

The Mg^{2+} whole rock concentration profile, Fig. 7b, exhibits two distinct depth trends. Between 95 mbgl and 155 mbgl, Mg^{2+} decreases from 1.7 mg g^{-1} to 1.2 mg g^{-1}. Below about 155 mbgl, whole rock Mg^{2+} is relatively high and much more variable with maximum values of about 3.0 mg g^{-1}. The Sr^{2+} profile shows no clear depth trend, except below 260 mbgl, where the concentrations are relatively high and constant at about 1 mg g^{-1}.

Discussion

PHYSICAL STRATIFICATION OF THE AQUIFER

The trends in matrix porosity and permeability illustrated in Fig. 4 reflect the variations in lithology with depth. Above 155 mbgl, the core samples consist predominantly of soft white Chalk; below 155 mbgl, samples contain incipient solution seams, irregular to planar clay-rich seams, and stylolites. This is illustrated in Fig. 4a where closed circles denote samples with marl seams and stylolites. These lithological features are indicative of localized pressure solution, and the highly variable matrix porosity and gas permeability below 155 mbgl are consistent with diagenetic unmixing due to pressure solution (Garison & Kennedy 1977, Scholle 1977, Scholle *et al.* 1983). Consequently, it is inferred that, above 155 mbgl, the dominant mode of diagenesis of the Chalk at Fair Cross was mechanical compaction but, below 155 mbgl, the matrix has been significantly modified by pressure solution processes. The absence of any spatial correlation in matrix porosity above 155 m, and the presence of a strong spatial correlation below 155 mbgl are also consistent with diagenetic unmixing in the deeper zone.

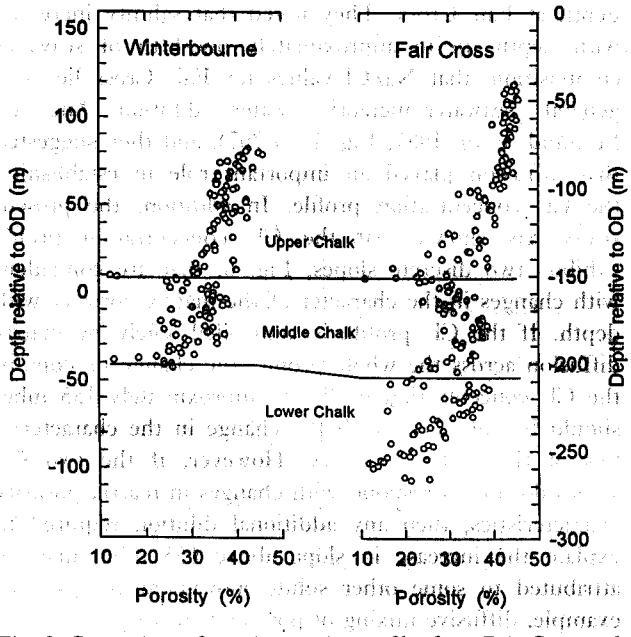

Fig. 8 *Comparison of matrix porosity profiles from Fair Cross and Winterbourne. The figure shows the positions of the Upper and Middle Chalk and the Middle and Lower Chalk boundaries in each of the boreholes. The two profiles have been centred on the Chalk Rock. Depths are in metres relative to OD.*

Evidence for the diagenetic boundary elsewhere in the confined Chalk is difficult to obtain as there are no equivalent data sets in the centre of the London Basin. However, a porosity profile for the unconfined Chalk at Winterbourne, previously described by Edmunds *et al.* (1973) and Bloomfield *et al.* (1995), can be compared with the Fair Cross porosity profile. The Winterbourne Borehole (NGR SU 4542 7161) is situated in the Winterbourne valley near Newbury, Berkshire, approximately 32 km north-west of Fair Cross (Fig. 2). Fig. 8 shows the Winterbourne and Fair Cross porosity profiles plotted on the same scale, centred on the top of the Middle Chalk. The Winterbourne borehole penetrates approximately 80 m of Upper Chalk, 45 m of Middle Chalk and terminates in the top few metres of the Lower Chalk. The sequence is relatively condensed with respect to Fair Cross, as illustrated by the Middle Chalk which is approximately 62 m thick at Fair Cross, but only 45 m thick at Winterbourne. Fig. 8 shows that the Winterborne porosity profile is broadly similar to that of Fair Cross; however, there is no evidence for two porosity populations associated with discrete depth intervals at Winterbourne. Porosities range from 12.5% to 45.9% and come from a single normal population with a mean of 33.7% (s.d. 6.4%). Samples from Winterbourne show only limited development of solution seams throughout the section and stylolites are present only in two samples from the bottom few metres of the section. The absence of a discrete change in diagenetic characteristics with depth at Winterbourne suggests that the diagenetically controlled porosity profile at Fair Cross may be representative only of the confined Chalk aquifer towards the centre of the London Basin.

On the basis of the regional hydrogeology and core data, a semi-quantitative model can be developed to describe the horizontal hydraulic conductivity profile at Fair Cross (Fig. 9). The profile between 75 and 155 mbgl has been calculated by assuming that the transmissivity in the confined Chalk in the vicinity of Fair Cross is of the order of 300 m^2d^{-1} (Owen & Robinson 1978) and that the effective aquifer thickness is 80 m, equivalent to the thickness of the Chalk above the zone of inferred pressure solution. At Fair Cross, flow may be concentrated in the highly fractured top 20 m of the Chalk, and Fig. 9 has been drawn assuming that 80% of the transmissivity is due to flow through this section. Below 155 mbgl, the horizontal hydraulic conductivity profile is based on the general variation in matrix permeability with depth. The model suggests that horizontal hydraulic conductivity at Fair Cross may be highly stratified with the upper section, between 75 and 155 mbgl, four to five orders of magnitude more conductive than the Chalk below 155 mbgl. Below 155 mbgl, the calculated hydraulic conductivity falls from 7.4×10^{-8} ms^{-1} at 155 mbgl to 1.5×10^{-9} ms^{-1} at the base of the Lower Chalk.

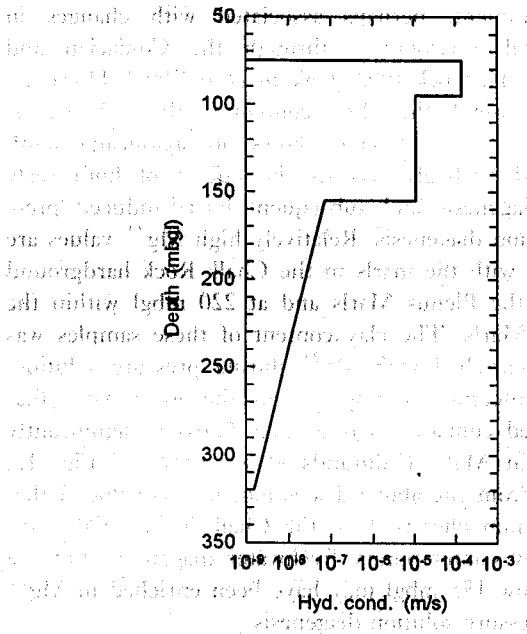

Fig. 9 *Illustration of the modelled horizontal hydraulic conductivity profile at Fair Cross. Depths are in metres below ground level.*

HYDROGEOCHEMICAL STRATIFICATION OF THE AQUIFER

Several of geochemical depth trends appear to correlate with changes in the physical properties of the Chalk at about 155 mbgl (Figs. 4, 6 and 7). The following section briefly describes some of the possible controls on the hydrogeochemistry at Fair Cross and discusses how depth trends in rock and water chemistry may relate to burial diagenesis and to contemporary hydrogeology processes.

Chemistry of the Chalk

The Mg^{2+} and Sr^{2+} contents of the carbonate are in the ranges 1.25 to 2.95 mg g^{-1} and 0.43 to 1.10 mg g^{-1}, respectively. These ranges are consistent with those previously reported for Chalk (Hancock 1975, Bath & Edmunds 1981). The Mg^{2+} content is broadly similar to or slightly higher than equivalent modern carbonate sediments (Sayles & Manheim 1975), but the Sr^{2+} values are relatively low. These low values are inferred to be primarily due to early marine diagenesis. This interpretation is consistent with the observation that low values of Sr^{2+} at approximately 205 mbgl and 265 mbgl are associated with the Chalk Rock and Melbourn Rock hardgrounds which probably underwent relatively prolonged early marine diagenesis. A plot of Sr^{2+} against Cl^- for pore waters from Fair Cross (Edmunds *et al.* 1987, *cf.* Fig. 12, p. 267) also indicates that evolved connate water is significantly enriched in Sr^{2+}. As the Chalk has a significant buffering capacity with respect to Mg^{2+} in the pore fluids, the Mg^{2+} composition of the rock predominantly reflects compositional changes in the original Chalk sediment, perhaps associated with changes in depositional environment through the Coniacian and Santonian (Hancock 1993, Jenkyns *et al.* 1994). However, below 155 mbgl, the Mg^{2+} content of the carbonate is higher, more variable and shows no significant depth trend, and probably records the effects of both early marine diagenesis and subsequent burial-induced pressure solution diagenesis. Relatively high Mg^{2+} values are associated with the marls in the Chalk Rock hardground complex, the Plenus Marls and at 220 mbgl within the New Pit Marls. The clay content of these samples was probably enriched with Mg^{2+} during pressure solution, and this inference is supported by the observations that the evolved connate water at Fair Cross is significantly depleted in Mg^{2+} (Edmunds *et al.* 1987, *cf.* Fig. 12, p. 267). From the above discussion, it is concluded that the strontium chemistry of the Chalk is essentially unaffected by contemporary freshwater diagenesis, but the Chalk below 155 mbgl may have been enriched in Mg^{2+} during pressure solution diagenesis.

Pore water chemistry

Edmunds *et al.* (1987, 1992) have previously described an overall increase in pore water mineralisation with depth at Fair Cross. They noted that salinity increased with depth up to approximately one-fifth of seawater composition, that Na/Cl values for Fair Cross lie on a general seawater–meteoric water dilution line (*cf* Edmunds *et al.* 1987, Fig. 12 p 267), and they suggested that diffusion played an important role in establishing the Cl^- concentration profile. In addition, the present study has shown that the Cl^- concentration profile exhibits two discrete slopes, Fig. 6, that are coincident with changes in the character of the matrix porosity with depth. If the Cl^- profile is controlled solely by matrix diffusion across the whole profile, the change in slope of the Cl^- concentration profile at approximately 155 mbgl should be consistent with the change in the characteristics of the matrix porosity. However, if the two Cl^- slopes do not correspond with changes in matrix porosity characteristics, then any additional dilution required to explain the increase in slope above 155 mbgl may be attributed to some other solute movement process, for example, diffusive mixing of pore and fracture waters.

If steady state conditions are assumed, then mass flux through the profile, $\mathcal{J}_{diff}$, is given by the expression

$$\mathcal{J}_{diff} = -D_E \, \delta c/\delta x = \text{constant} \qquad (1)$$

where $D_E = D_T\psi$. D_E is the effective diffusion coefficient, $\delta c/\delta t$ is the concentration gradient, D_T is the tracer (free water) diffusion coefficient and ψ is diffusibility. Diffusibility is given by the expression $\phi_D \, \delta/\tau^2$, where ϕ_D is the diffusive porosity, δ is constrictivity and τ^2 is tortuosity. As the ratio of the Cl^- concentration gradient between 95 to 155 mbgl to that below approximately 155 mbgl is approximately 2 (Fig. 6), under steady state conditions a difference of a factor of 2 in diffusibility is required if there is a steady state diffusive flux of Cl^- across the full profile.

Given that above 155 mbgl the average porosity is 44% and that below 155 mbgl it is 30% (Fig. 4), approximately half the difference in the slope of the concentration profile can be ascribed simply to a reduction in matrix porosity below 155 mbgl. A decrease in constrictivity associated with a decrease in matrix porosity will also contribute to the decreased slope below 155 mbgl, but some or all of this could be lost by a increase in the tortuosity. Although there are uncertainties in values of constrictivity, tortuosity and D_E (D_E may vary from approximately 0.5^{-10} to 3×10^{-10} m^2s^{-1} Hill 1984) it is inferred that the change in porosity across 155 mbgl is insufficient to account for all the observed increase in slope of the Cl^- concentration profile above 155 mbgl. It is therefore inferred that above 155 mbgl the Cl^- profile may be affected by processes such as diffusional exchange between matrix and fracture waters as well as matrix diffusion. However, it is unclear whether the assumption of steady state is reasonable. It would take approximately 2 million years for a 200 m profile to

approach equilibrium under steady state conditions (Bath & Edmunds 1981, Barker 1991), and the effect of even very small hydraulic gradients on the chloride profile over such a time-scale is uncertain.

The Na^+ concentration profile shows an increased mineralisation with depth, (Fig. 6b). It broadly parallels trends in Cl^- concentration. However, there is a disparity in the change in the slope of the Cl^- and the Na^+ concentration gradients. This is probably due to the relative enrichment of Na^+ in the pore fluids above 155 mbgl, either through cation exchange with clays in the Chalk matrix (Ineson & Downing 1963, Downing *et al.* 1979, Edmunds *et al.* 1987, 1992) and/or through leakage of Na^+-rich waters from the overlying Palaeogene sands and clays. Unlike the Cl^- profile, the Na^+ profile extends throughout the full sequence of the Chalk at Fair Cross. Below approximately 260 mbgl, corresponding to the Lower Chalk (Fig. 3), the Na^+ concentration is approximately constant at 1800 mgl^{-1}. From this it is inferred that the Lower Chalk may act as the end member for diffusive mixing with fresh waters entering the top of the profile. Since the Plenus Marls and the Melbourn Rock at the top of the Lower Chalk consist of some of the least porous units in the entire Chalk sequence, these marls and hardgrounds may have an important role in regulating the overall solute concentration profiles at Fair Cross.

The concentrations of Mg^{2+} and Sr^{2+} in pore waters at Fair Cross increase with depth (Fig. 7a and 7c), and the profiles are associated with a general increase in salinity. In both profiles, the greatest rate of increase is in the 95 to 155 mbgl interval. For example, below 155 mbgl, the rate of Mg^{2+} increase is approximately one fifth of that above 155 mbgl. Depth profiles of Mg/Ca and Sr/Ca molar ratios for Fair Cross, (Fig. 10), indicate that contemporary incongruent dissolution of the carbonate is occurring between 95 and 155 mbgl but, below 155 mbgl, it is inferred that the Chalk is not undergoing contemporary freshwater diagenesis.

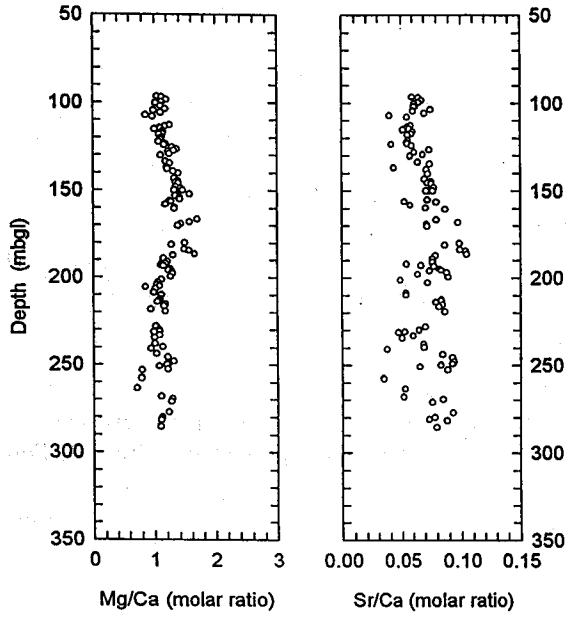

Fig. 10 *Illustration of the variation in (a.) molar Mg/Ca and (b.) molar Sr/Ca ratios with depth at Fair Cross, where depths are in metres below ground level.*

PALAEOHYDROGEOLOGY OF THE AQUIFER

Four principal stages in the post-Albian development of the Chalk aquifer of the London Basin can be identified (Anderton *et al.* 1979); A. initial Chalk sedimentation and early marine diagenesis during the Cenomanian to Campanian, B. sub-aerial erosion between the end of the Campanian and the early Palaeogene, C. burial during the Palaeogene, and D. renewed uplift between the end of the Palaeogene and the present. Despite this general framework, it is difficult to constrain the timing of the major phase of pressure solution at Fair Cross. This is because there are significant uncertainties in i) the maximum thicknesses of Chalk and Palaeogene sediments that were deposited in the London Basin, and ii) the evolution of the chemistry of the pore waters. It is not clear whether the Chalk overburden was sufficient to generate pressure solution during the Cenomanian to Campanian, or whether pressure solution was induced by the Palaeogene phase of burial.

In southern England, the maximum preserved thickness of the Chalk is about 500 m in Hampshire and a maximum preserved thickness of about 450 m has been reported for the English Channel (Rawson *et al.* 1978). In addition, studies of Tertiary uplift and Chalk exhumation histories from southern UK off-shore basins have suggested that up to approximately 1 km of Chalk overburden was removed in these areas during the Tertiary (Hillis 1991, 1995, Smith *et al.* 1994). Consequently, although the original maximum thickness of Chalk at Fair Cross is uncertain, it is assumed that there was at least 400 to 500 m of Chalk deposited at Fair Cross. If 500 metres of Chalk were deposited, the position of the diagenetic boundary, presently at 155 mbgl, would have been at a depth of about 330 m. The maximum preserved thickness of Palaeogene sediments in the Thames valley is 320 m, in the western English Channel it is about 400 m, and in the Isle of Wight it is about 600 m (Curry *et al.* 1978). Again, although the original maximum thickness of Palaeogene overburden at Fair Cross is uncertain, if there were 400 m of Palaeogene sediments originally deposited at Fair Cross then the diagenetic boundary would have been at a depth of 480 m. The burial depth at which solution transfer becomes an important process is poorly defined because it is a function of the pore water chemistry. However, if the Chalk

at the end of the Cretaceous contained connate pore waters but by the time of Palaeogene burial, these had been diluted by fresher groundwaters, then given the minimum likely Chalk and Palaeogene overburdens described above, it follows that the major phase of pressure solution diagenesis at Fair Cross occurred during Palaeogene burial, at depths below about 400 to 500 m, and not during the late Cretaceous. An additional observation that supports this inference is that the Winterbourne borehole, which lies away from the known Palaeogene depositional centre of the London Basin, does not show the same diagenetic transition as that at Fair Cross.

On the basis of the above discussion, a schematic representation of some aspects of the palaeohydrogeology of the Chalk at Fair Cross is presented in Fig. 11. Porosity and Cl^- pore water concentration profiles are used to illustrate possible general trends in the physical and chemical evolution of the aquifer. It is emphasized that the profiles in Fig. 11 are intended only to provide a basis for discussion and are not intended to be definitive descriptions of the variation in porosity or chloride concentration with time.

By the end of the Campanian, about 400 to 500 m of Chalk had probably been deposited (column A of Fig. 11). It is envisaged that the matrix porosity profile was similar to profiles seen in contemporary pelagic carbonate oozes and chalks (Schlanger & Dougals 1974), *ie.* decreasing from approximately 60% at the sediment-water interface to about 30 to 40% at a burial depth of approximately 400 m. The chloride concentration is assumed to have approximated to seawater concentration throughout the full sequence. Sub-aerial exposure and erosion of the Chalk throughout the London Basin is inferred from the unconformity between the top of the Chalk and the Palaeogene. Column B in Fig. 11 illustrates possible depth trends in porosity and chloride concentration in the Chalk at Fair Cross before Palaeogene sedimentation. If it is assumed that the matrix porosity

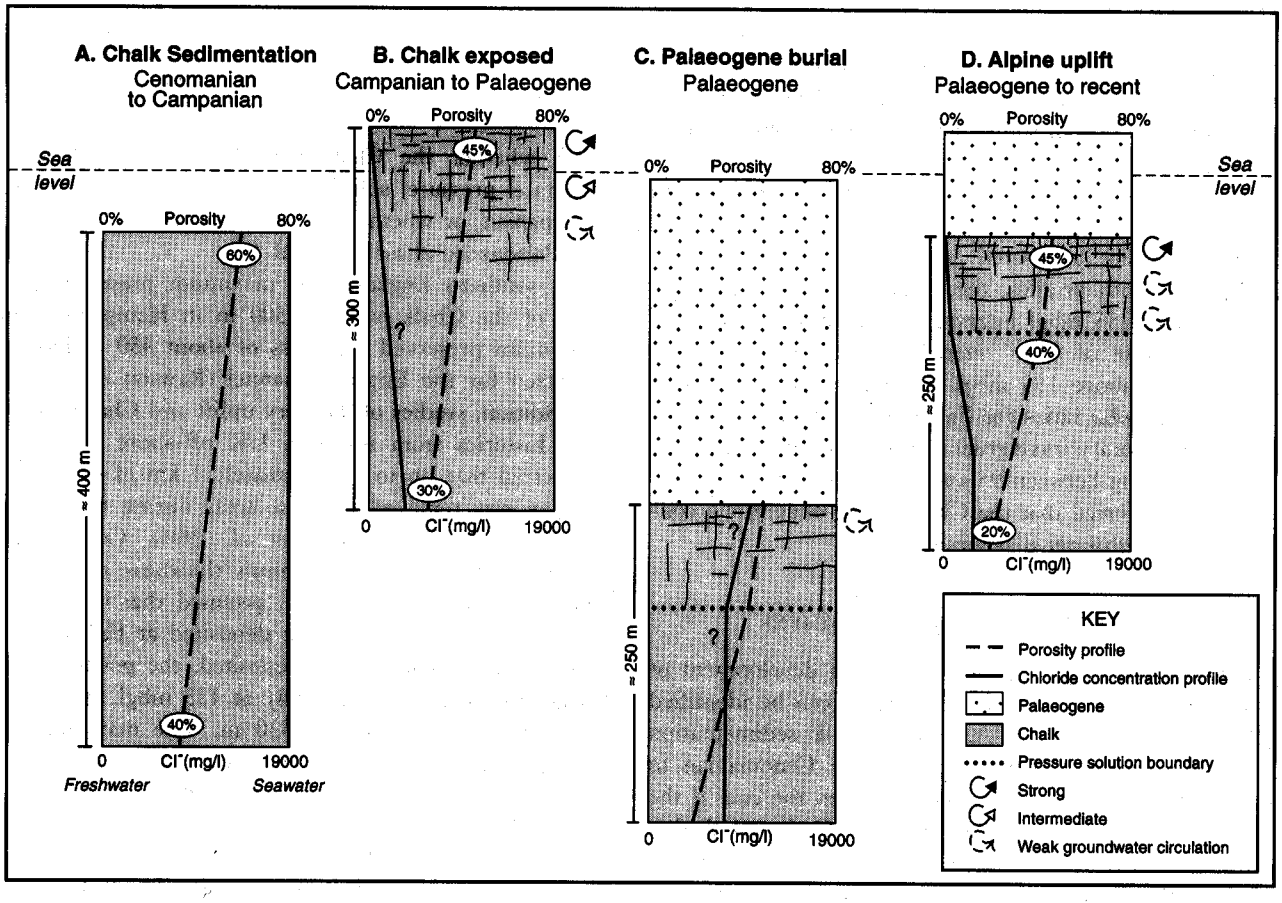

Fig. 11 *Schematic illustration of the four principal stages in the evolution of the confined Chalk aquifer at Fair Cross and the inferred variation in porosity and chloride depth profiles with time. A. initial Chalk sedimentation and early marine diagenesis during the Cenomanian to Campanian, B. sub-aerial erosion between the end of the Campanian and the early Palaeogene, C. burial during the Palaeogene, and D. renewed uplift between the end of the Palaeogene and the present. Inferred porosity and Cl^- matrix concentration profiles are denoted by dashed and heavy solid lines respectively, in addition, intervals of fracturing and inferred groundwater circulation are schematically represented on the figure. The position of the mechanical compaction—pressure solution boundary is indicated by the dotted line.*

profile was predominantly controlled by mechanical compaction, then porosity was likely to range from about 40 to 45% at the top of the profile to approximately 30% at the base of the profile. Unloading of the Chalk during sub-areal weathering would have led to the development of fracturing in the top of the section, but it is not known to what extent the fracture system had developed by the start of the Palaeogene. Pore waters at the top of the profile would have been diluted to a composition approaching that of the fresh water recharge. At the base of the Chalk, a maximum salinity of about one fifth seawater may be expected, equivalent to present values at the base of the Chalk, but the salinity could have been substantially less.

By the end of the Palaeogene the Chalk at Fair Cross may have been buried by a few hundred metres of Palaeogene deposits. Column C in Fig. 11 illustrates the possible porosity and chloride concentration depth trends at the end of Palaeogene sedimentation. During this period, the Chalk was buried to a sufficient depth that the lower part of the profile underwent a significant degree of pressure solution diagenesis (the diagenetic transition is indicated by the horizontal dotted line). Above the zone of pressure solution, there were likely to have been negligible changes in porosity; however, within the zone undergoing pressure solution, porosities were reduced significantly and became increasingly variable as pressure solution progressed. Following pressure solution diagenesis, porosities as low as 20% were probably typical at the base of the Chalk. It is assumed that groundwater circulation was greatly reduced during Palaeogene burial. Fracture apertures are expected to have been reduced with the increased overburden, and one possible effect of pressure solution would have been to close fractures through cementation in the zone of pressure solution. During Palaeogene burial, water entering the upper part of the profile would have been increasingly saline, although it should be noted that the Reading Beds are a non-marine facies and that, at least initially, there was the potential for continued freshwater recharge to the top of the Chalk. It is not known whether pressure solution was maintained throughout the Palaeogene, however the water in the lower section of the profile would have had to remain relatively fresh, or at least relatively depleted in Mg^{2+}, while pressure solution proceeded. If Mg^{2+} were lost from the pore water and incorporated into clays during pressure solution diagenesis, the decrease in the Mg^{2+} concentration of the deep pore waters may have been sufficient to maintain pressure solution in an increasingly saline environment. However, it is extremely difficult to constrain the possible extent of mixing between the relatively fresh pore waters and the assumed increasingly saline recharge water by the end of the Palaeogene. Consequently, the position of the Cl^- profile in column C, Fig. 11, is even more arbitrary than in the other three profiles.

Between the end of Palaeogene sedimentation and the present, the London Basin underwent Alpine uplift, primarily during the Miocene. Column D in Fig. 11 illustrates graphically some aspects of the present Chalk profile at Fair Cross. The present porosity profile is probably essentially unchanged from the one that developed during Palaeogene burial and effectively reflects the maximum burial depth of the sediments (Hillis 1995). Groundwater circulation is essentially limited to the Chalk unaffected by pressure solution. However, it is likely that groundwater circulation in the top 20 to 80 m of the Chalk at Fair Cross has been enhanced significantly since the end of the Palaeogene. Younger (1989) has suggested that cooler groundwaters during the Devensian may have caused solution enlargement of pre-existing fracture networks in the Chalk and glacial episodes throughout the Quaternary would have reduced the Palaeogene cover substantially, increasing the potential for freshwater recharge to the centre of the Basin. The development of enhanced conductivity in the upper section of the confined Chalk at Fair Cross may also have been associated with the establishment of preferential flow paths throughout the London Basin and the development of the Lower Thames valley as an area of discharge (Lloyd 1993).

Conclusions

1. At Fair Cross, the matrix porosity of the Chalk above 155 mbgl is relatively high and constant at approximately 40 to 43%. Below 155 mbgl, porosity is greatly reduced and more variable. This change in the matrix porosity characteristics is controlled by a change in the dominant mode of burial diagenesis, *ie.* from mechanical compaction above 155 mbgl to compaction by pressure solution below 155 mbgl. The present matrix porosity profile has probably remained essentially unchanged since it was established during the Palaeogene, when the Chalk experienced a maximum overburden.
2. Comparison of the matrix porosity profile from the confined Chalk at Fair Cross with an equivalent porosity profile for the unconfined Chalk at Winterbourne, Berkshire, suggests that the diagenetic boundary developed only in the deep confined Chalk of the London Basin and is not present at the margins of the Basin.
3. Four hydrogeologically distinct depth intervals can be recognized at Fair Cross. The basal interval, between 267 and 322 mbgl, consists of the Lower Chalk. Matrix porosity is low and variable, fractures are absent and there is probably negligible flow through this interval. Salinities are constant with depth and are relatively high (Na^+ 1800 mgl^{-1}). Pore waters in this interval may act as a source of salinity for diffusive mixing with fresh waters entering the top of the

profile and the Plenus Marls and the Melbourn Rock at the top of the Lower Chalk may have an important role in regulating the overall concentration profiles of solutes at Fair Cross.

4. In the interval between 155 and 265 mbgl, porosities are low but highly variable due to pressure solution diagenesis. There is no pervasive fracturing and there is probably negligible flow through this section of the profile. Any fractures present prior to Palaeogene burial may have been cemented by pressure solution processes and the relatively compacted and cemented matrix probably inhibited the subsequent development of fractures during Alpine uplift. Solute concentration profiles in this interval are inferred to be primarily a function of matrix diffusion.
5. The depth interval between 95 and 155 mbgl lies above the zone of Chalk affected by pressure solution diagenesis. Porosities are high and are controlled primarily by mechanical compaction. Fracturing is present throughout the interval but it is probably preferentially developed near the top of the interval. Increases in pore water Mg/Ca and Sr/Ca molar ratios with depth indicate that the Chalk is undergoing incongruent dissolution and that there is contemporary groundwater circulation in this interval. An increase in the chloride concentration gradient above 155 mbgl cannot be attributed solely to an increase in the matrix porosity and it is inferred that, in addition to matrix diffusion, the solute profiles above 155 mbgl are affected by diffusive exchange between matrix and fracture waters.
6. The upper depth interval, between 74 and 95 mbgl, is pervasively fractured. It is inferred that this unit corresponds to the zone of enhanced hydraulic conductivity reported from the top of the confined Chalk of the London Basin. Solute movement in this zone is probably by advection and by diffusive exchange between matrix and fracture waters.
7. The Chalk at Fair Cross must have been extensively flushed with relatively fresh water by the end of the post-Cretaceous uplift to have enabled pressure solution to occur towards the base of the Chalk during the Palaeogene. The fracture network at the top of the present Chalk sequence was probably initiated during the late-Cretaceous and early-Palaeogene uplift, but it is likely that the network was enhanced significantly since the start of the Quaternary with erosion of the cover.
8. This study has shown that if a more detailed understanding of the hydrogeology of carbonate aquifers, such as the Chalk, is to be developed, it is essential that the full geological and diagenetic history of the aquifer is considered. This is especially important in the confined Chalk where the effects of recent weathering are less likely to dominate the contemporary hydrogeology.

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